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New many-particle system consisting of electron pairs and holes in a two-dimensional crystal

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When atoms are organized into a crystal, the single-electron energy levels of individual atoms form energy bands. However, there also exist electron-pair states in atoms. We found previously that the counterpart of these electron-pair states of individual atoms may survive in a two-dimensional square-lattice crystal. When the amplitude of the crystal potential is larger than a certain value, a metastable electron-pair band arises between the two lowest single-electron energy bands. In this paper, we demonstrate that the many-particle exchange-correlation corrections renormalize the energy bands and stabilize electron pairs for certain hole and electron-pair densities in the two-dimensional crystal. Therefore, depending upon the crystal potential and free carrier densities, there may exist a new many-particle system consisting of electron pairs and holes. Furthermore, we calculate the charge-density excitation spectrum in this new many-particle system.

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Electronic band-structure in solid-state theory is based on single-electron states. It is known that the single-electron energy levels of periodically organized individual atoms form energy bands in a crystal. As a matter of fact, individual electron pairs also exist in atoms and ions, such as the negative hydrogen ion H^- and helium atom He, etc.[1, 2]. The quantum state of an electron pair is different in its nature from the single-electron state because of the strong correlation between the two paired electrons. Locality of the electron correlation is essential in the electron-pair states, where the correlation reduces greatly the Coulomb repulsion between them. The electron pair in an atom localizes closely to the positive charge of the nucleus and does not form a valence with other atoms. In our previous work[3], we show that the counterpart of the electron-pair states of individual atoms may survive in a two-dimensional (2D) square lattice crystal. When the amplitude of the periodic potential of the crystal is larger than a certain value, a metastable electron-pair band arises between the two lowest single-electron energy bands at low electron density limit. This metastable state represents two correlated electrons of spin singlet bound in the same unit cell in relative coordinates. The wavefunctions of two paired electrons are of maximum overlap each other in the crystal and, consequently, strong correlation between them reduces greatly their Coulomb repulsion. Based on the single electron and electron-pair band structure, we have also developed a Hamiltonian of a many-particle system with electron-pairs and single electrons[4] in the 2D system. Two different scenarios were considered for the single-electron band below the electron-pair band. It can be the lowest conduction band or the highest valence band of a real crystal.

When the single-electron band is assumed as the highest valence band of a crystal, the conduction carriers in this band are holes with charge $+e$, effective mass m^*, and dispersion relation $E^h_{\mathbf{k}}$. The crystal is intrinsically an insulator. The valence band is fully occupied by electrons at zero temperature and the hole density is zero. The metastable electron-pair band stays above this valence band with an energy gap $E_g^{(0)}$ at zero density limit[3]. The primary goal of this work is to demonstrate that these metastable electron-pair states can be stabilized by including many-particle exchange-correlation energy in such a 2D system.

We assume that there are N_{hole} holes in the valence band and N_{pair} electron pairs (with effective mass $2m^*$ and charge $-2e$) in the electron-pair band. The total carrier charge number in the system is given by $N_0 = N_{hole} - 2N_{pair}$. Transition of two electrons from the valence band to the electron-pair band creates an electron pair leaving behind two holes in the valence band, or vice verse[4]. The scenario is schematically shown in Fig. 1. Our calculations below will show that in the presence of free electron pairs and holes, the many-particle exchange-correlation effects renormalize the energy gap and can indeed stabilize the electron-pair states. Therefore, there may exist a ground state of a new two-dimensional many-particle system consisting of electron pairs and holes in a two-dimensional crystal depending upon the crystal po-

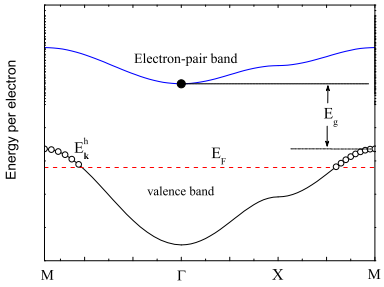

FIG. 1: A schematic plot of the energy band structure with electron pairs and holes in a 2D square lattice.

tential and free carrier densities.

Taking the top of the valence band as the reference for energy, the many-particle Hamiltonian in this system can be written as[4],

$$\tilde{H} = \tilde{H}_0 + \tilde{H}_1, \tag{1}$$

with

$$\begin{aligned}\tilde{H}_0 = & \sum_{\mathbf{k},\sigma} E^h_{\mathbf{k}} d^\dagger_{\mathbf{k},\sigma} d_{\mathbf{k},\sigma} + \sum_{\mathbf{k}} \left(E^{\rm pair}_{\mathbf{k}} + 2E^{(0)}_g \right) b^\dagger_{\mathbf{k}} b_{\mathbf{k}} \\ & + \frac{1}{2A} \sum_{\mathbf{k_1},\mathbf{k_2},\mathbf{q}} \sum_{\sigma,\sigma'} v_q d^\dagger_{\mathbf{k_1}-\mathbf{q},\sigma} d^\dagger_{\mathbf{k_2}+\mathbf{q},\sigma'} d_{\mathbf{k_2},\sigma'} d_{\mathbf{k_1},\sigma} \\ & + \frac{1}{2A} \sum_{\mathbf{k_1},\mathbf{k_2},\mathbf{q}} v_{pp}(q) b^\dagger_{\mathbf{k_1}-\mathbf{q}} b^\dagger_{\mathbf{k_2}+\mathbf{q}} b_{\mathbf{k_2}} b_{\mathbf{k_1}} \\ & + \frac{1}{A} \sum_{\mathbf{k},\mathbf{k_1},\mathbf{q},\sigma} v_{hp}(q) b^\dagger_{\mathbf{k_1}-\mathbf{q}} d^\dagger_{\mathbf{k}+\mathbf{q},\sigma} b_{\mathbf{k_1}} d_{\mathbf{k},\sigma}, \end{aligned} \tag{2}$$

and

$$\tilde{H}_1 = \frac{1}{\sqrt{A}} \sum_{\mathbf{k},\mathbf{q}} v^{\rm t}(q) \left(b^\dagger_{\mathbf{k}} d^\dagger_{\frac{\mathbf{k}}{2}+\mathbf{q},\uparrow} d^\dagger_{\frac{\mathbf{k}}{2}-\mathbf{q},\downarrow} + b_{\mathbf{k}} d_{\frac{\mathbf{k}}{2}-\mathbf{q},\downarrow} d_{\frac{\mathbf{k}}{2}+\mathbf{q},\uparrow} \right), \tag{3}$$

where the operators $d^\dagger_{\mathbf{k},\sigma}$ and $d_{\mathbf{k},\sigma}$ are creation and annihilation operators, respectively, for a hole of momentum $\hbar\mathbf{k}$ and spin σ. They obey the fermion anti-commutation relations $\{d_{\mathbf{k},\sigma}, d^\dagger_{\mathbf{k}',\sigma'}\} = \delta_{\mathbf{k},\mathbf{k}'}\delta_{\sigma,\sigma'}$, $\{d_{\mathbf{k},\sigma}, d_{\mathbf{k}',\sigma'}\} = 0$, and $\{d^\dagger_{\mathbf{k},\sigma}, d^\dagger_{\mathbf{k}',\sigma'}\} = 0$. The operators $b^\dagger_{\mathbf{k}}$ and $b_{\mathbf{k}}$ are creation and annihilation operators, respectively, for an electron pair of momentum $\hbar\mathbf{k}$ and spin zero. They obey the commutation relations $[b_{\mathbf{k}}, b^\dagger_{\mathbf{k}'}] = \delta_{\mathbf{k},\mathbf{k}'}$, $[b_{\mathbf{k}}, b_{\mathbf{k}'}] = 0$, and $[b^\dagger_{\mathbf{k}}, b^\dagger_{\mathbf{k}'}] = 0$. In this work, we will use the units of effective Bohr radius $\mathrm{a}_B = \epsilon_0\hbar^2/m^*e^2$ and effective Rydberg $\mathrm{R}_y = \hbar^2/2m^*\mathrm{a}_B^2$ for length and energy, respectively. For a crystal with effective electron mass $m^* = 5m_0$ and static dielectric constant $\epsilon_0 = 30$, one obtains $\mathrm{a}_B = 3.17$ Å and $\mathrm{R}_y = 75.6$ meV. As discussed in Ref.[4], the hole-hole interaction potential v_{hh} is the same as that of the single electron-electron interaction giving by $v_{hh} = v_{ee} = v_q = 2\frac{2\pi}{q}$. The electron-pair-electron-pair [hole-electron-pair] interaction potential is given by $v_{pp}(q) = 4v_q f_{pp}(q)$ [$v_{hp}(q) = -2v_q f_{ep}(q)$] with a form factor $f_{pp}(q) \leq 1$ [$f_{ep}(q) = \sqrt{f_{pp}(q)}$]. The second part $\tilde{H}_1$ of the total Hamiltonian is responsible for creating (destructing) an electron pair in the electron-pair band and two holes in the valence band at the same time and $v^t(q)$ is the interacting potential. The interaction potentials $v_{pp}(q)$, $v_{hp}(q)$, and $v^t(q)$, as well as the form factors are given in Ref. [4].

In order to stabilize the electron pairs and simplify the calculations, we will first consider the hole density n_{hole} as an input and take the electron pair density as $n_p = n_{hole}/2$. This means that we assume the total carrier charge number $N_0 = 0$ (it can also be taken as a constant). In this way, $\tilde{H}_1$ will not be invoked in present calculations. However, $\tilde{H}_1$ may be important for further studies on the dynamic processes or temperature effects in the system. The calculations in this work are performed for zero temperature. The exchange-correlation energies of the electron pairs and holes will be obtained as a function of the hole and electron-pair densities.

Thus, we are dealing with a two-component plasma consisting of holes (fermions) and electron pairs (bosons) in a 2D crystal. Within the linear response theory, the density-density response function is given by[5, 6],

$$\{[\chi(q,\omega)]^{-1}\}_{ij} = [\chi^{(0)}_{ii}(q,\omega)]^{-1}\delta_{ij} - \varphi_{ij}(q), \tag{4}$$

for $i, j = p$ (pair) and h (hole), where $\chi^{(0)}_{ii}(q,\omega)$ is the non-interacting polarizability of the ith component and $\varphi_{ij}(q)$ is the static effective interaction potential between the components i and j. The effective potential $\varphi_{ij}(q)$ defines a local field correction in terms of the "bare" potential $v_{ij}(q)$. Within the random-phase approximation (RPA), the local field correction on the static effective interaction is neglected and therefore $\varphi_{ij}(q) = v_{ij}(q)$.

The many-particle interaction energy in such a system of boson-fermion mixture can be obtained by[7]

$$E_{ij} = \int_0^{e^2} \frac{E^{int}_{ij}(\lambda)}{\lambda} d\lambda, \tag{5}$$

where the inter-particle interaction potential $E^{int}_{ij}(\lambda)$ depends on the static structure factor, given by

$$E^{int}_{ij}(e^2) = \frac{1}{2A} \sum_{\mathbf{q}} v_{ij}(q)[S_{ij}(q) - \delta_{ij}]. \tag{6}$$

The static structure factor can be calculated by[7],

$$S_{ij}(q) = -\frac{1}{\pi\sqrt{n_i n_j}} \int_0^\infty d\omega \chi_{ij}(q, i\omega). \tag{7}$$

For a non-interacting 2D boson (electron-pair) gas with density n_p at zero temperature, we can assume that all the electron pairs are in the same state at the bottom of the electron-pair band, i.e., in the condensate phase[6, 8]. The polarizability of the non-interacting boson gas is given by,

$$\chi^{(0)}_{pp}(q, i\omega) = -\frac{2n_p\varepsilon_{q,p}}{\omega^2 + \varepsilon^2_{q,p}}, \tag{8}$$

where $\varepsilon_{q,p} = q^2/2$. For a non-interacting 2D fermion (hole) gas with density n_h, we have[9, 10]

$$\chi^{(0)}_{hh}(q, i\omega) = -\frac{1}{4\pi\varepsilon_q}\{2\varepsilon_q - \sqrt{2}[a + \sqrt{a^2 + (2\varepsilon_q\omega)^2}]^{\frac{1}{2}}\}, \tag{9}$$

where $a = \varepsilon_q^2 - 4\varepsilon_q\varepsilon_{_F} - \omega^2$, $\varepsilon_q = q^2$, $\varepsilon_{_F} = E_0 - E_F$ and E_0 is the top of the valence band ($\varepsilon_{_F} > 0$).

If we take $v_{hp}(q) = 0$, i.e., without considering hole-electron-pair interaction, the static structure factor of the boson gas of electron pairs within the RPA is given by

$$S_{pp}(q) = [1 + 2n_p v_{pp}(q)/\varepsilon_{q,p}]^{-\frac{1}{2}}. \tag{10}$$

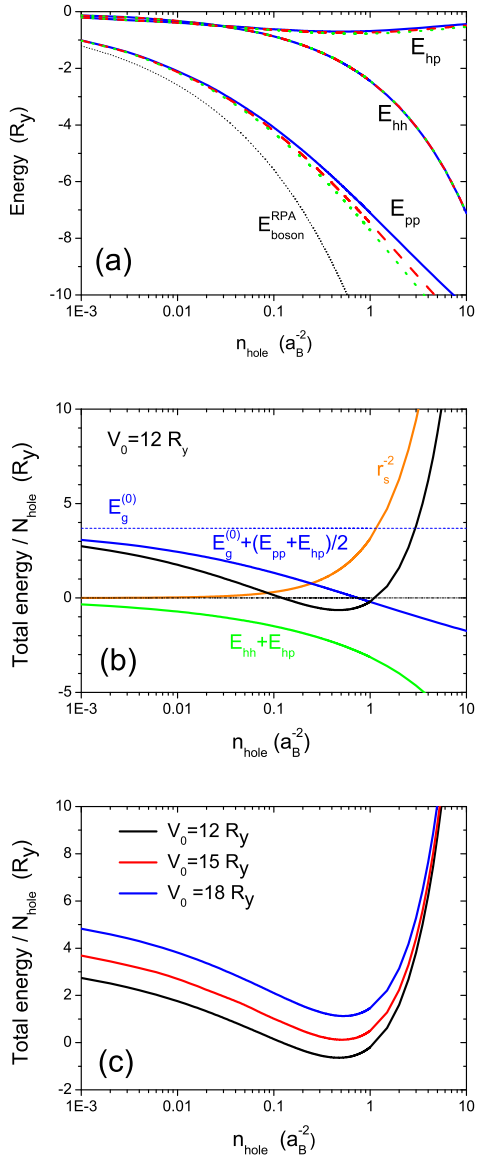

FIG. 2: (a) The many-particle interaction energies E_{pp}, E_{hh}, and E_{hp} as a function of hole density in the 2D square lattices with $\lambda = 1.5$ a$_B$ and $V_0 = 12$ (blue solid curves), 15 (red dash curves), and 18 R$_y$ (green dotted curves). The thin-dotted curve is the RPA result for an ideal 2D boson gas. (b) Total energy (black solid curve) of the interacting electron pairs and holes in the 2D square lattice of $V_0 = 12$ R$_y$. The contributions of hole kinetic energy (orange curve), hole (green curve) and electron-pair (blue curve) exchange-correlation energies are also shown. The horizontal blue dashed line indicates $E_g^{(0)}$. (c) A comparison of the total energies for different lattice potentials $V_0 = 12$, 15, and 18 R$_y$. $\lambda = 1.5$ a$_B$.

Consequently, the pair-pair interaction energy becomes,

$$E_{pp} = 8(r_s^p)^{-\frac{2}{3}} I, \qquad (11)$$

where $r_s^p = (\pi n_p)^{-\frac{1}{2}}$, and

$$I = \int_0^\infty dx\{(x^3/4)[\sqrt{1+8f_{pp}(q)/x^3}-1]-f_{pp}(q)\}. \quad (12)$$

For $f_{pp}(q) \equiv 1$ (i.e., considering the bosons as point charges), the above integral becomes $I = I_0 = -1.29$. This is the well-known RPA result for charged boson gas[11]. Notice that the factor 8 in Eq. (11) is due to the units used here.

Following our previous calculations[3, 4], we will consider a 2D square lattice crystal with an effective periodic potential $V_c(x,y) = V_0[\cos(2\pi x/\lambda)+\cos(2\pi y/\lambda)]$ with period λ and amplitude V_0. Especially, for $\lambda = 1.5$ a$_B$ and $V_0 = 12$, 15, and 18 R_y, we have obtained $E_g^{(0)} = 3.69$, 4.64, and 5.78 R$_y$, respectively[3]. Fig. 2(a) shows the many-particle interaction energies E_{pp}, E_{hh}, and E_{hp} obtained using Eqs. (5-9) within the RPA in such 2D square lattices. The electron-pair density is taken as $n_p = n_{hole}/2$ in the calculation. The hole-hole interaction energy E_{hh} is very close to the RPA energy of a 2D charged fermion gas[12] with a small difference resulting from the hole-electron-pair coupling. The pair-pair energy E_{pp} depends on V_0, especially at high density, because the crystal potential affects the size of the electron pair (the average distance between two paired electrons). In the figure, we also give the RPA result of an ideal 2D charged boson gas[11] $E^{\rm RPA}_{\rm boson} = -8 * 1.29(r_s^p)^{-2/3}$. The difference between E_{pp} and $E^{\rm RPA}_{\rm boson}$ is due mainly to finite size (against point charge) of the electron pairs and pair-hole coupling. We now can estimate the total energy of the system as shown in Fig. 2(b). Taking the top of the valence band as zero energy, the total energy of N_{hole} holes in the valence band is given by $E_{hole} = N_{hole}(r_s^{-2} + E_{hh} + E_{hp})$ with $r_s = 1/\sqrt{\pi n_{hole}}$, where the first term is the kinetic energy of the holes. The total energy of N_{pair} ($= N_{hole}/2$) electron pairs in the electron-pair band is given by $E_{pair} = N_h[E_g^{(0)} + (E_{pp} + E_{hp})/2]$. The total energy of the system is given by $E_{tot} = E_{hole} + E_{pair}$. The competition of the kinetic and exchange-correlation energies leading to a minimum of the total energy of the system. In Fig. 2(c) we compare the total energies in the square lattices ($\lambda = 1.5$ a$_B$) of different potentials $V_0 = 12$, 15, and 18 R$_y$, where the minimum of the total energy occurs at n_{hole} =0.473, 0.508, and 0.525 a$_B^{-2}$ with values -0.641, 0.123, and 1.127 R$_y$, respectively.

The presence of free carries in a crystal leads to renormalization of the band structures. It is well known that exchange-correlation corrections of free carriers reduce the band gap in semiconductor materials. It was found that the exchange-correlation energy is almost independent of band characteristics both in 3D and 2D electron-hole plasmas in semiconductors. The many-particle exchange-correlation energy depends only on the interparticle distance r_s (or particle density) in appropriate rescaled natural units in a universal manner[13–15]. The band-gap renormalization (BGR) can be obtained by calculating the average exchange-correlation energy per

particle or by the self-energies of the particles involved. The kinetic energy is usually assumed to be unchanged in a renormalization process.

Using the above obtained interaction energies, we can estimate the BGR in the present system with an electron-pair band and a hole valence band. The many-particle interactions reduce the energies of an electron pair and a hole by quantities $\Delta_p = (E_{pp}+E_{hp})$ and $\Delta_h = E_{hh}+E_{hp}$, respectively. Therefore, the energy gap between the two bands is reduced by $\Delta E_g = \Delta_h+\Delta_p/2$ as shown in Fig. 3. As soon as the bottom of the electron-pair band touches the Fermi energy E_F of the valence band, the electron pairs cannot decay into two single electrons (or cannot annihilate with two holes) in the valence band because of the exclusion principle. Therefore, the electron pairs are stabilized. Our calculation results in Fig. 3 indicate that this happens for $n_{hole} \geq n_c = 0.108$, 0.244, and 0.570 a_B^{-2} in the 2D crystals with $V_0 = 12$, 15, and 18 R_y, respectively. For hole densities larger than n_c, the electron pairs become stable with density $n_p = n_{hole}/2$. We also notice that the half-filling of the valence band in the present system occurs at $n_{half} = 1/\lambda^2 = 0.444$ a_B^{-2}. The critical density $n_c < n_{half}$ for $V_0 = 12$ and 15 R_y and $n_c > n_{half}$ for $V_0 = 18$ R_y. Calculations beyond the RPA will enhance the correlation energy and consequently reduce the hole density n_c. Therefore, the present RPA results show definitely the existence of a new 2D many-particle system consisting of electron pairs and holes with certain stable densities of electron pairs. Notice that, for higher density $n_{hole} \geq n_c$, the present calculation for band energies is not valid. Quantitative results of the band energies require a self-consistent calculation considering the charge redistribution in the two band system. However, one may expect the bottom of the electron-pair band stays closely to the Fermi level of the valence band for high density because of the bosonic characteristic of the electron pairs.

As soon as the electron pairs are stabilized, we can discuss the elementary excitations in this two component plasma system. The collective excitation modes of charge-density waves can be obtained by solving the equation $\det|\chi^{-1}(q,\omega)| = 0$. The dynamic structure factor of this many-particle system is given by

$$S_{ij}(q,\omega) = -\frac{1}{\pi\sqrt{n_i n_j}}\mathrm{Im}\chi_{ij}(q,\omega). \qquad (13)$$

Fig. 4(a) shows the plasmon dispersions (the black solid curves) obtained within the RPA for a two-component plasma with hole density n_{hole}=0.15 a_B^{-2} slightly larger than n_c and electron-pair density $n_p = n_{hole}/2 = 0.075$ a_B^{-2} in the 2D square lattice of $\lambda = 1.5$ a_B and $V_0 = 12$ R_y. We also show in the figure the plasmon dispersions of a hole gas of density n_{hole}=0.15 a_B^{-2} (green dashed curve) and an electron-pair gas of density n_p=0.075 a_B^{-2} (orange dashed curve) without hole-electron-pair interaction (i.e., taking $v_{hp}(q) \equiv 0$). It is known that, for both 2D fermion and boson gases,

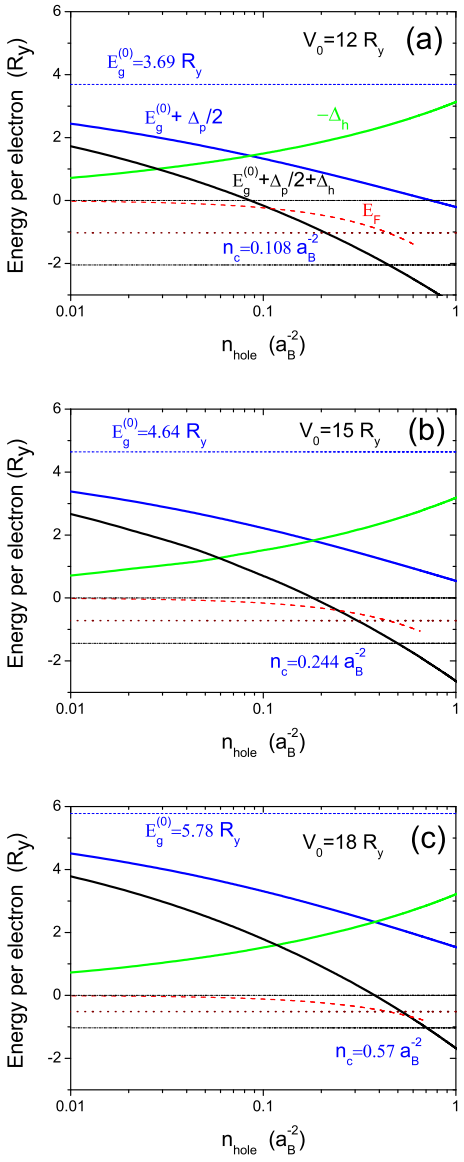

FIG. 3: Band gap renormalization (BGR) in the 2D square lattices with $\lambda = 1.5$ a_B and V_0 = (a) 12, (b) 15, and (c) 18 R_y. The energy gap $E_g^{(0)}$ is indicated by the horizontal blue-dashed line. With increasing the hole and electron-pair densities, the electron-pair band is lowered by $\Delta_p/2$ (blue-solid curve) and the valence band is elevated by $-\Delta_h$ (green-solid curve). The energy gap is reduced by $\Delta_p/2+\Delta_h$. Taking the top of the valence band as $E = 0$, the valence band is in between the two dotted-dash horizontal lines and the Fermi level E_F is given by the red-dashed curve. The renormalized bottom of the electron-pair band is given by the black-solid curve and it touches the Fermi level at $n_{hole} = n_c$. The half-filling of the valence band occurs at $n_{hole} = \lambda^{-2} = 0.444$ a_B^{-2} where E_F is at the middle (the brown dotted line) of the valence band.

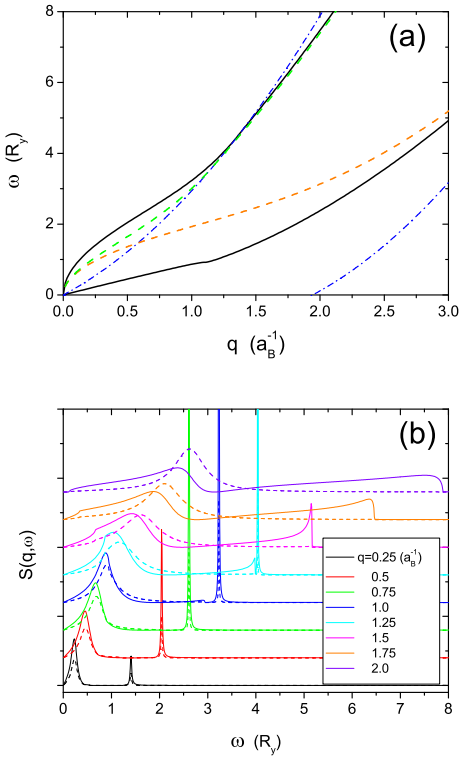

FIG. 4: (a) The dispersions of charge-density excitations of the two-component plasma consisting of electron pairs (bosons) and holes (fermions) with n_{hole}=0.15 a_B^{-2} and $n_p = n_{hole}/2$ in the 2D square lattice of $\lambda = 1.5$ a_B and $V_0 = 12$ R_y. The green-dashed and orange-dashed curves are the plasmon dispersions of the hole and electron-pair gas, respectively, without including hole-electron-pair interaction. The hole single-particle continuum is in between the two blue dotted-dash curves. (b) The corresponding dynamic structure factor $S_{hh}(q,\omega)$ (the solid curves) and $S_{pp}(q,\omega)$ (the dash curves) for different wavevectors.

the plasmon frequency is proportional to $\sqrt{q}$ at long-wavelength limit ($q \to 0$) as is confirmed by the green and orange dashed curves in Fig. 4(a). In this case, the hole plasmon (the green dashed curve) is Landau damped in the hole single-particle continuum. In contrast to the Fermion gas of holes, there is no Landau damping within the RPA for the boson gas plasmon of the electron-pairs. When hole-electron-pair interaction is taken into account, two branches of the plasmon modes (the two black curves) are found. The hole-electron-pair interaction alters significantly the plasmon modes of the system. Our calculations show that, at long wavelength limit, the lower branch of the charge-density excitation modes is proportional to q and the higher one to $\sqrt{q}$. The lower branch is localized in the hole single-particle excitation continuum and the high one is above the up-limit of the hole single-particle continuum for $q \lesssim 1.3$ a_B^{-1} in this case. In order to understand better the charge-density excitations, we have calculated the dynamic structure factor as shown in Fig. 4(b). Because the holes and electrons pairs are strongly coupled to each other, the structure factors $S_{hh}(q,\omega)$ and $S_{pp}(q,\omega)$ are quite similar when the higher branch of the plasmon mode is above of the hole single-particle continuum (for $q \lesssim 1.3$ a_B^{-1}). We observe that, for $q = 0.25$, 0.5, 0.75, 1.0, and 1.25 a_B^{-1}, both $S_{hh}(q,\omega)$ and $S_{pp}(q,\omega)$ show two well defined peaks in Fig. 4(b). The low-frequency peak is of certain width and the higher frequency one (above the hole single-particle continuum) shows a δ function (a small broadening is artificially introduced to show the δ-function peak). Therefore the dynamic structure factors demonstrate that, though the lower branch of the plasmon mode is localized in the single-particle continuum, it is only partially damped because of its partial bosonic origin. For higher wavevectors (i,.e, $q = 1.5$, 1.75 and 2.0 a_B^{-1}), where the higher branch of the plasmon mode is in the single particle continuum region of the hole gas, $S_{hh}(q,\omega)$ and $S_{pp}(q,\omega)$ become very different. For $q = 2.0$ a_B^{-1}, e.g., $S_{hh}(q,\omega)$ shows the single-particle spectrum of holes and $S_{pp}(q,\omega)$ shows a broad peak corresponding to a partially damped collective mode of electron-pair gas due to hole-electron-pair coupling.

In conclusion, we have shown that there exists a new two-dimensional many-particle system of boson-fermion mixture consisting of electron pairs and holes in 2D square lattice crystals. We calculated the ground-state energy of this many-particle system within the RPA and analysed the band-gap renormalization. We also obtained the collective charge-density excitation spectrum of such a boson-fermion mixture. We hope that the theoretical results obtained in this work can enhance our understanding on electronic properties of crystals.

Acknowledgments

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